

# Linear Temperature Dependence of the Lower Critical Field $H_{c1}$ in F-Doped LaOFeAs Superconductors

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We present the first experimental results of the lower critical field  $H_{c1}$  of the newly discovered F-doped superconductor  $\text{LaO}_{0.9}\text{F}_{0.1}\text{FeAs}$  (F-LaOFeAs) by global and local magnetization measurements. It is found that  $H_{c1}$  showed an clear linear- $T$  dependence down to a temperature of 2 K, indicative of an unconventional pairing symmetry with a nodal gap function. Based on the  $d$ -wave model, we estimated a maximum gap value  $\Delta_0 = 4.0 \pm 0.6$  meV, in consistent with the recent specific heat and point-contact tunneling measurements. Taking the demagnetization factor into account, the absolute value of  $H_{c1}(0)$  is determined to be about 54 Oe, manifesting a low superfluid density for  $\text{LaO}_{0.9}\text{F}_{0.1}\text{FeAs}$ .

PACS numbers: 74.20.Rp, 74.25.Ha, 74.70.Dd

The recent discovery of novel superconductivity in rare-earth iron-based layered superconductors has received great attention in the scientific community [1, 2, 3, 4, 5, 6, 7, 8, 9, 10, 11, 12, 13, 14, 15, 16]. Except for the cuprate superconductors, these new materials  $\text{ReO}_{1-x}\text{F}_x\text{FeAs}$  exhibit quite high critical temperatures: 26-28K for Re=La at  $x=0.05$ -0.11[1, 2, 3, 4], 36K for Re=Gd at  $x=0.17$ [10], 41K for Re=Ce with  $x=0.16$ [7], 43K for Re=Sm with  $x=0.15$ [6], and 52K for Re=Nd, Pr at  $x=0.15$ [8, 9], as well as 25K in hole doped  $\text{La}_{1-x}\text{Sr}_x\text{OFeAs}$  [5]. One of the crucial issues to understand the underlying superconducting mechanism in such transition metal-based systems is the pairing interaction, *i.e.*, the symmetry of the superconducting order parameter and the nature of the low energy excitations. Up to now, there have been several investigations on the pairing symmetry for  $\text{LaO}_{0.9}\text{F}_{0.1}\text{FeAs}$  (F-LaOFeAs) superconductor. Recent specific heat measurement has revealed a nonlinear magnetic field dependence of the electronic specific heat coefficient  $\gamma(H)$  in the low temperature limit, which is consistent with the prediction for a nodal superconductor [17]. The presence of a node in the superconducting gap has also been verified by the observation of a zero-biased conductance peak in point contact tunneling spectroscopy [18]. On the other hand, an extended  $s$ -wave multi-band superconductivity was proposed [11] to account for the experiment results of the extremely high upper critical field in F-LaOFeAs. From the band structure point of view, the two-band model has been strengthened with  $d$ -wave pairing interactions originated from the intra-band antiferromagnetic coupling plus an effective inter-band antiferromagnetic interaction [16]. Because the current experiments have been performed on polycrystalline samples, the experimental results within the context of the symmetry of the pair state is not yet well determined.

Lower critical field  $H_{c1}(T)$ , or magnetic penetration depth  $\lambda(T)$  are fundamental probes of the existence of nodes, or two-gap in the superconducting gap function of unconventional superconductors. As an advantage,  $H_{c1}(T)$  measurement probe relatively large distances ( $\lambda \sim 1000$  Å) and are far less sensitive to sample surface quality. In this work we present the first detailed measurements of the temperature dependence of

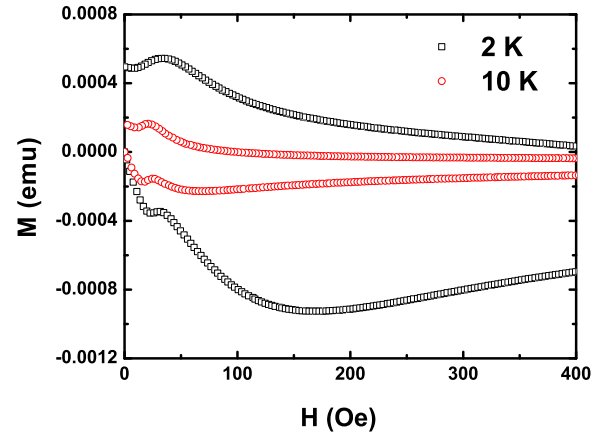


FIG. 1: (Color online) Magnetization hysteresis loops of  $\text{LaO}_{0.9}\text{F}_{0.1}\text{FeAs}$  measured by VSM in ZFC mode at  $T = 2, 10$  K.

the lower critical field  $H_{c1}$  of superconducting  $\text{LaO}_{0.9}\text{F}_{0.1}\text{FeAs}$  polycrystals. We found a predominately linear- $T$  dependence of  $H_{c1}$  down to a temperature of 2 K, which is a strong evidence for nodes in the superconducting gap of F-LaOFeAs samples.

Our polycrystalline samples were synthesized by using a two-step solid state reaction method. The superconducting transition temperature  $T_c$  defined as the onset of the drop in resistivity was 26 K with a transition width of  $\Delta T_c = 3$  K (10%-90% of normal state resistivity). The details of the sample synthesis and the characterization have been reported in our previous papers [3, 17]. Our samples were relatively dense with a metallic shiny grain surface and shaped into various dimensions. The data presented here were taken on a platelike polycrystal with the dimension of  $0.6 \times 0.6 \times 0.2$  mm<sup>3</sup>.

Global dc magnetization measurement were carried out by a high resolution vibrating sample magnetometer (VSM) (Quantum Design) with applied field  $H$  parallel to the shortest lateral side of the sample. Local magnetization loops were

measured using a two dimensional electron gas (2DEG) micro Hall probe with an active area of  $10 \times 10 \mu\text{m}^2$ . The Hall probe was characterized without sample attachment at different temperatures. In our experiment, all  $M(H)$  curves were taken in zero field cool (ZFC) mode with initial temperature up to 40 K. To minimize the complex effects of the character of the field penetration in layered structure, such as Bean-Livingston surface barriers and/or geometrical barriers [19, 20], we used a low field sweep rate of 30 Oe/min to measure isothermal magnetization curves.

Typical global magnetization loops  $M(H)$  of our specimen at  $T=2$  and 10 K are presented in Fig. 1. It is seen that the  $M(H)$  curves are symmetric and show a relatively large critical current density in the mixed state using Bean critical state model. This implies that the surface barriers for flux entry does not play an important role in our case since Bean-Livingston surface barriers are expected to give rise to a very asymmetric magnetization loop [21], and the pinning mechanism mainly arises from bulk pinning. In addition, a feature of a peak in  $M(H)$  appears followed the full penetration field. This so called “second peak” in  $M(H)$  loops has been widely observed in strongly layered superconductors, such as  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$  and  $\text{NdCeCuO}$  single crystals, and attributed to the vortex phase transition to vortex glass state [22, 23]. Here the observation of second peak effect in our specimen indicates that the  $\text{LaO}_{0.9}\text{FeAs}$  is strongly anisotropic and the intergrain links are strong, representative of the bulk property.

Figure 2(a) and Figure 2(b) are the initial isothermal  $M(H)$  curves measured by VSM and Hall probe over the temperature range from 2 to 20 K, respectively. The second peak effect is more distinct in the curves by local measurement, as shown in Fig. 2(b). These  $M(H)$  curves show clearly a linear dependence of the magnetization on field caused by Meissner effect at low fields. In global measurement, due to the bulk pinning the entry of vortices is gradual, comparing to the local measurement. This causes a gradual  $M(H)$  curve, as shown in Fig. 2(a). It is difficult to determine  $H_{c1}$  using these  $M(H)$  curves. Therefore we use the method in Ref. 24 to extract the value of  $H_{c1}$ : According to the Bean critical state model for type-II superconductors, magnetic induction  $B$  induced by the external field is:

$$B = A(H - H_{c1})^2/H^* \quad (H_{c1} < H < H^*), \quad (1)$$

where  $A$  is a sample-dependent constant and  $H^*$  is related to critical current density. According to this model, a plot of  $B^{1/2}$  vs  $H$  should yield a straight line with a threshold at  $H_{c1}$ . Fig. 3 shows such plot of  $B^{1/2}$  vs  $H$  at  $T = 2, 3, 4$  and 10 K for the global curves, where  $B^{1/2} = (M - M_{ML})^{1/2}$  and  $M_{ML}$  is the Meissner line for each curve, shown as the dotted line in Fig. 2 for the curve of  $T = 2$  K. As displayed, the data are well described by Eq. (1) except a small kink at  $H > H_{c1}$ . This kink is due to the second peak effect in the original  $M(H)$  curves. Neglecting this effect,  $H_{c1}$  is well determined as the field of the threshold of  $B$ , as demonstrated in Fig. 3. By the same procedure, we also determined the  $H_{c1}(T)$  data from the

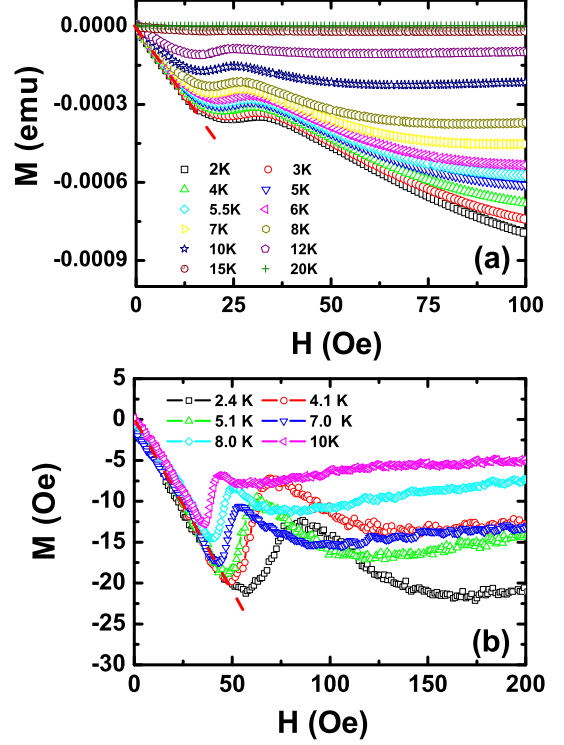


FIG. 2: (Color online) The magnetization curve  $M(H)$  by VSM (a) and Hall probe (b), at various temperatures, respectively. The dotted red lines are the “Meissner line” showing the linearity of these curves at low fields.

$M(H)$  curves using Hall probe (not shown here for simplicity).

Depicted in Fig. 4(a) are our main results, in which the extracted  $H_{c1}$  is plotted as a function of  $T$  for both VSM and Hall probe measurements. As shown in the inset of Fig. 4(a), the  $H_{c1}$  measured by VSM is much lower than those by local measurement even though we measured the same sample. The reason for this difference is as follows. In global measurement, the VSM coil picks up any signal once vortices start to enter into the specimen through the edge of the sample. On the other hand, the local Hall probe detects signal only when vortices almost fully penetrates into the sample and reaches the region where the Hall probe locates. Assuming the Hall probe is located at the center of the sample, which is far from the sample edge, the Hall probe detects signal only from the central region. Accordingly, the  $H_{c1}$  measured by local Hall sensor is expected to be larger than that of global VSM measurement. It also explains the experimental results that the  $M(H)$  curves by global measurement is more gradual around the penetration region, comparing those measured by local magnetization measurement, which is exactly the case we observed in Fig. 2. The fact that the demagnetization factor is different for local and global measurements can manifest itself

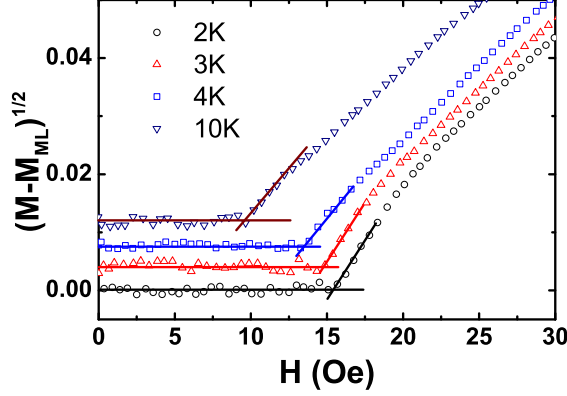


FIG. 3: (Color online) The  $M(H)$  data in the form of  $(M - M_{ML})^{1/2}$  vs  $H$ .  $M_{ML}$  is the Meissner line shown in Fig. 2. The curves ( $T=3,4,10$  K) are shifted up for clarity. The colored lines are guides to the eyes.

by scaling the  $H_{c1}(T)$  curve measured by VSM to the one by Hall sensor. After we correct a scaling factor of 3.22 for the  $H_{c1}(T)$  by VSM, the two  $H_{c1}(T)$  lines almost collapse onto the same line except the part of high  $T$ , which is due to large error bars, as shown in the main panel of Fig. 4(a). The coincidence of the two scaled  $H_{c1}(T)$  curves indicates that our results are intrinsic in physics, independent of measuring device and technique. Thereafter, if not specially mentioned, we choose the data obtained by VSM for our analysis and discussions since the demagnetization factor is well determined for global measurement, as discussed below.

We now analyze and discuss the obtained data of  $H_{c1}(T)$ . Principally one can get the absolute value of  $H_{c1}(0)$  by extrapolating the nominal  $H_{c1}(T)$  curve to  $T = 0$ , taking the demagnetization factor into account. For samples with elliptical cross sections,  $H_{c1}$  can be deduced from the first penetration field  $H_{c1}^*$ , assuming that the magnetization  $M = -H_{c1}$  when the first vortex enters into the sample. Thus  $H$  has been rescaled to  $H = H_a - NM$  and  $H_{c1} = H_{c1}^*/(1 - N)$ , where  $N$  is the demagnetization factor and  $H_a$  the external field. For sample with a rectangle cross section, the  $M(H)$  curves can still be approximately by a linear variation in the Meissner state. However, the demagnetization factor has been reconsidered. It has, for instance, been shown by Brandt that a bar with a rectangle cross section, the first penetration field  $H_{c1}^*$  and  $H_{c1}$  have the relation [25]:  $H_{c1} = H_{c1}^*/\tanh(\sqrt{0.36b/a})$ , where  $a$  and  $b$  are the width and the thickness of the samples, respectively. Using this formula, we estimate the effective demagnetization factor  $N_{eff} \approx 0.67$  as we take  $a \approx 0.6$  mm and  $b \approx 0.2$  mm for our sample. This estimated  $N_{eff}$  is consistent with those of  $\text{MgB}_2$  platelets with the similar size ratio [26, 27], in which  $N_{eff} \approx 0.6 - 0.69$  has been determined.  $H_{c1}^*(0) = 18$  Oe is obtained by linear fitting to the data of  $H_{c1}(T)$  below 5 K. With  $N_{eff}$ , the formula  $H_{c1}(0) = H_{c1}^*(0)/(1 - N_{eff})$  yields a true  $H_{c1}(0) = 54$  Oe. A striking feature of the nominal  $H_{c1}(T)$

shows an approximate linear- $T$  dependence from around 6 K down to 2 K, the lowest temperature we can reach in this experiment, as shown by the straight line in the inset of Fig. 4(a). This approximate linear- $T$  behavior is also evidenced from the  $H_{c1}(T)$  curve using Hall sensor.

In order to investigate the properties of superconducting gap using the obtained linear- $T$  dependence of  $H_{c1}$  in the low- $T$  region, we use the formula to calculate the absolute value of the penetration depth  $\lambda$  (taking the demagnetization effect into account):

$$H_{c1} = \frac{\Phi_0}{4\pi\lambda^2} (\ln \kappa + 0.5), \quad (2)$$

where  $\Phi_0 = hc/2e = 2.07 \times 10^{-7}$  Oe  $\text{cm}^2$  is the flux quantum, and  $\kappa$  is the Ginzberg-Laudau parameter. Assuming  $\kappa \sim 100$ , we deduce a value of  $\lambda \approx 390$  nm with the calculated parameter  $H_{c1}(0) = 54$  Oe.

In Fig. 4(b) we plot the deviation  $\Delta\lambda = \lambda(T) - \lambda(0)$  as a function of  $T$  using  $\lambda(0) = 390$  nm. At low temperatures  $T < 5$  K,  $\Delta\lambda$  shows a linear function of  $T$ , i. e.  $\Delta\lambda \propto T$ , which is more explicit in the inset of Fig. 4(b). This linear dependence of  $\Delta\lambda$  contradicts the result of an isotropic  $s$ -wave gap superconductivity. For an isotropic  $s$ -wave superconductor, such as conventional superconductor, the low excitation of the finite energy gap is a type of an exponentially thermal activation. In the isotropic  $s$ -wave BCS theory,  $\Delta\lambda = \lambda(0) \sqrt{\pi\Delta_0/2T} \exp(-\Delta_0/T)$ , where  $\Delta_0$  is the maximum gap value. This theory can not describe our  $\Delta\lambda(T)$  since the exponential term gives  $d\lambda(T)/dT \approx 0$  at low  $T$ , shown as the fitting line in the inset of Fig. 4(b), in contrast to our result of  $d\lambda(T)/dT \propto T$ .

The linear- $T$  behavior of  $\Delta\lambda$  has been observed in high- $T_c$  superconductor YBCO [24, 28],  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$  [29, 30] single crystals,  $\text{HgBa}_2\text{Ca}_2\text{Cu}_3\text{O}_{8+\delta}$  crystal powder [31] and magnetic superconductor  $\text{YNi}_2\text{B}_2\text{C}$  [32]. In cuprate superconductors, such a linear- $T$  dependence of penetration depth  $\lambda$  at low  $T$  is a signature for the existence of a line node in the superconducting energy gap function, i.e. the  $d$ -wave pairing [28, 31], where thermal excitation of quasiparticles near nodes in the superconducting energy gap cause  $\rho_s \equiv 1/\lambda^2$  to decline linearly with temperature. Based on the  $d$ -wave pairing theory,  $H_{c1}(T)/H_{c1}(0)$ , i.e.,  $\lambda^2(0)/\lambda^2(T)$ , is a linear- $T$  function at  $T \ll T_c$ , and can be expressed as [31, 33]:

$$1 - \frac{H_{c1}(T)}{H_{c1}(0)} = 1 - \frac{\lambda^2(0)}{\lambda^2(T)} = 2 \ln 2 \frac{k_B T}{\Delta_0}. \quad (3)$$

As shown in Fig. 4(a), the slope of the fitting line  $d(H_{c1}(T)/H_{c1}(0))/dT$  is 0.78, which yields a  $\Delta_0 = (1.8 \pm 0.3)k_B T_c$  based on Eq. (3). Using the known  $T_c = 26$  K, we found  $\Delta_0 = 4.0 \pm 0.6$  meV. This deduced  $\Delta_0$  are in excellent agreement with the values obtained by our recent specific heat [17] and point-contact tunneling spectroscopy [18] measurements on similar samples.

Before conclusion, we would like to address the concern of the effect of impurity scattering on the linear- $T$  dependence

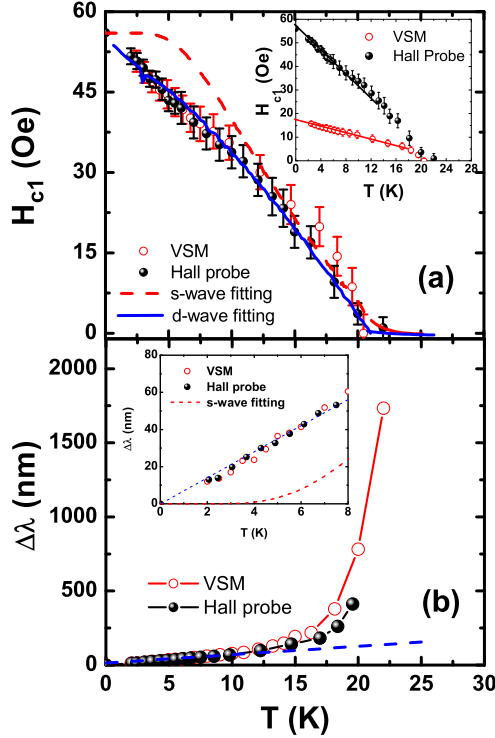


FIG. 4: (Color online) (a)  $T$  dependence of  $H_{c1}$  determined from the points deviating from the linearity of the initial  $M(H)$  curves by VSM and Hall Probe measurements. The data of  $H_{c1}(T)$  by VSM is scaled by multiplying a number of 3.22 to account for the different demagnetization effects. The solid blue line is a  $d$ -wave weak-coupling BCS fit with  $\Delta_0 = 3.85$  meV; the dashed red line is an  $s$ -wave weak-coupling fit. Inset: the original data of  $H_{c1}(T)$  measured by VSM and Hall Probe, respectively. The solid lines are the linear fit to the data of low  $T$ . (b)  $T$  dependence of  $\Delta\lambda$  calculated with the data of  $H_{c1}(T)$  and Eq. (2). Inset: the enlarged low temperature part, which shows a clear linear- $T$  dependence of  $\Delta\lambda$ . The dotted blue line is the linear fit, and the dashed red line is an  $s$ -wave exponential- $T$  line.

of magnetic penetration depth. For our samples, the coherence length  $\xi_0 \sim 80$  Å [17] and the mean free path  $l > 100$  Å [3], and thus our samples are in the moderate clean limit ( $l \sim \xi_0$ ). Meanwhile, we believe that nonmagnetic impurity scattering does not strongly modify the low- $T$  properties of F-LaOFeAs since  $T_c$  of our sample, another important low- $T$  parameter, does not change much while impurity level (residual resistivity) increases up to 2-4 times [3]. This situation also manifests itself by the fact that although different groups adopt different methods to synthesize F-LaOFeAs superconductor with different impurity level, the  $T_c$  defined as the drop of resistance is around 25-28 K [1, 4, 7, 11, 17].

To summarize, we conduct both global and local magnetization measurements on the new superconductor LaO<sub>0.9</sub>F<sub>0.1</sub>FeAs. The temperature dependence of the lower critical field  $H_{c1}$  is extracted. It is found that  $H_{c1}$  show a linear- $T$  behavior at low temperatures, suggesting a nodal

gap function. We also obtained the lower critical field of  $H_{c1}(0) = 54$  Oe, showing a low superfluid density for F-LaOFeAs. Based on the finite slope of  $H_{c1}(T)$  curve at low  $T$ , we estimated the maximum gap value to be  $\Delta(0) = 4.0 \pm 0.6$  meV, which is very consistent with the results of recent specific heat and tunneling experiments on the similar samples.

Acknowledgement: The authors are grateful to Prof. Wen-Xin Wang and Prof. Jun-Ming Zhou for providing GaAs/AlGaAs substrates. This work is supported by the National Science Foundation of China, the Ministry of Science and Technology of China (973 project No: 2006CB60100, 2006CB921107, 2006CB921802), and Chinese Academy of Sciences (Project ITSNEM).

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